Production of highly concentrated intermediate molybdenum isotopes in optimal cascade with two additional product flows

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Abstract. The problem of highly concentrated molybdenum isotopes production in multiflow cascades is considered. A method of the cascade with two additional product flows optimization has been developed. Two components intermediate in weight are concentrated simultaneously in those flows. The optimization problem is solved using a variation of the partial flow cuts of the cascade stages with large separation coefficients. The optimization criterion is the minimum of the stage total feed flow while ensuring a given concentration of isotopes. A computational experiment was carried out to separate a mixture of molybdenum hexafluoride. The experiment demonstrated the features of intermediate components concentrating in additional product flows of the cascade.

1. Introduction

Highly concentrated molybdenum isotopes $^{92,\ 100}$Mo, being the extremum on mass number, are recommended to use for a production of reactor materials with improved thermophysical properties [1, 2]. Enrichment of such isotopes, at the concentration close to 100 %, can be made in triple-flow ordinary cascades. Molybdenum isotopes, which are intermediate in mass between $^{94\text{-}98}$ Mo, can be used in other fields. Enrichment of such components in ordinary cascades is difficult and limited to the extreme values [3]. The mixture depleted of isotopes $^{94\text{-}97}$ Mo, which is received concurrently, can be used in reactor materials because of reduced cross section of neutron absorption. One way for solution of the problem is a synthesis of highly concentrated intermediate isotope in additional product in a long multi-flow cascade. Such isotope can be received from the stage with the highest concentration. Efficiency of the given method is shown on the *R* and *Q* model cascades with the variable feed flow through the stages as well as on the rectangular sectionalized cascades with the identical flow through particular sections of stages [4-9]. Differently concentrated isotopes can be as well received in the cascade with two additional products [10-11].

The purpose of the present paper is exploration of details of cascades optimization with concurrent concentration of intermediary components in two additional products. The calculations were carried out using the common method of calculation of multi-flow cascades [10]. Optimization was performed by the variation of partial flows' cuts at high splitting ratio of stages, which correspond to gas centrifuges. Consideration of cascades optimization with two additional products was made by a computing experiment by separation of molybdenum hexafluoride mixture.

2. Parameters and equations of multi-flow cascade

The design model of multi-flow double-ended cascade contains *n* stages (figure 1). The cascade consists of feed flow, main product, where the lightest isotope is enriched, and waste flow, where the heaviest isotope is enriched. The same characteristics are used for the product and waste stages. Each stage has additional product flows of cascade. Additional product flows are taken from the main product and waste flows of stages which differ in isotopes composition. The scheme makes it possible to calculate cascades with any set of external flows. This asks that all the missing flows have to be equal to zero.

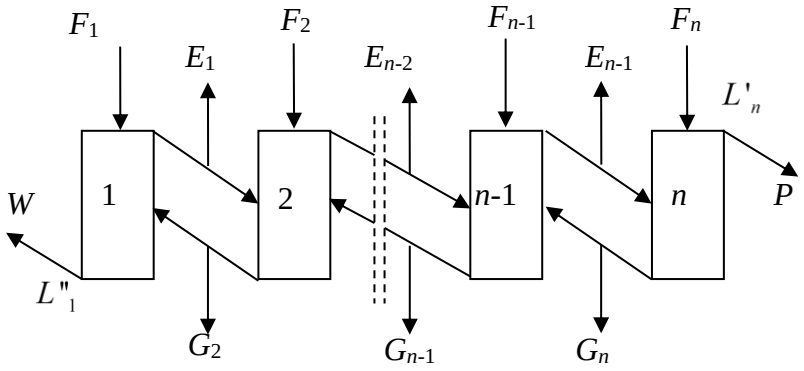

Figure 1. The calculation scheme of a multi-flow cascade: 1, 2, *n*-1, *n* – stage number.

Figure 1 shows the agreed notations of external flows of cascade: W – waste, F_1, F_2,…, F_n – feed, P – main product, E_1, E_2,…, E_{n-1} and G_2, G_3,…, G_n – additional products from the select and waste flows of stages consequently. Let's indicate concentration (molar fraction) of *j*-component in flows: C_j^W – waste, $C_{j,1}^F, C_{j,2}^F, ..., C_{j,n}^F$ – feed, C_j^P – main product, $C_{j,1}^E, C_{j,2}^E, ..., C_{j,n-1}^E$ and $C_{j,2}^G, C_{j,3}^G, ..., C_{j,n}^G$ – additional products, $j = \overline{1,m}$. Then at loss-free condition, the equations of matter and each component balance in a cascade are true.

$$\sum_{i=1}^{n} F_i = P + W + \sum_{i=1}^{n-1} E_i + \sum_{i=2}^{n} G_i;$$

$$\sum_{i=1}^{n} F_i C_{j,i}^F = PC_j^P + WC_j^W + \sum_{i=1}^{n-1} E_i C_{j,i}^E + \sum_{i=2}^{n} G_i C_{j,i}^G, \quad j = \overline{1,m}.$$

Component-wise sum of concentrations for any flow equals to one. Accordingly, from the given equations there are *m* independent.

Internal parameters of cascade include feed, product and waste flows of stages – L_i, L'_i, L''_i. and concentrations of components in these flows – $C_{j,i}, C'_{j,i}, C''_{j,i}$ $i = \overline{1,n}$, $j = \overline{1,m}$. They are linked by the equations of matter and component balance, written for each stage in a similar way to the cascade formula.

In the given scheme each stage with number $i = \overline{2,n-1}$ feeds by the flow $L'_{i-1} – E_{i-1}$ of the previous and $L''_{i+1} – G_{i+1}$ subsequent stages. At the ends of the cascade to the first stage is fed the flow $L''_2 – G_2$, to the last $L'_{i-1} – E_{i-1}$. In addition the stage has an external feed flow F_i. From here it follows that the equations of constraint of stages flows are expressed as balance correlations

$$L_1 = L_2'' - G_2 + F_1; \quad L_n = L'_{n-1} - E_{n-1} + F_n; \quad L_i = L'_{i-1} - E_{i-1} + L''_{i+1} - G_{i+1} + F_i;$$

$$L_1 C_{j,1} = L_2'' C''_{j,2} - G_2 C_{j,2}^G + F_1 C_{j,1}^F; \quad L_n C_{j,n} = L'_{n-1} C'_{j,n-1} - E_{n-1} C_{j,n-1}^E + F_n C_{j,n}^F;$$

$$L_i C_{j,i} = L'_{i-1} C'_{j,i-1} - E_{i-1} C^E_{j,i-1} + L''_{i+1} C''_{j,i+1} - G_{i+1} C^G_{j,i+1} + F_i C^F_{j,i},\ i = \overline{2, n-1},\quad j = \overline{1, m}.$$

The separation characteristics of stages are set by the total ratios of splitting $q_{i,j}$, $j = \overline{1, m}$, $i = \overline{1, n}$. Let's number the components in the order of increasing of their mass number and take the heaviest isotope as the base component. Then

$$q_{j,i} = \frac{C'_{j,i} C''_{m,i}}{C''_{j,i} C'_{m,i}}.$$

In the molecularly-kinetic methods of separation the total ratios of splitting $q_{i,j}$ are shown through $\Delta M_{jm} = M_m - M_j$ according to the formulas $q_{j,i} = \left(q_{0,i}\right)^{\Delta M_{jm}}$, where $q_{0,i}$ – splitting ratio per unit of mass number difference.

Taking into consideration the boundary conditions which link the external and internal parameters

$$W = L''_1;\ P = L'_n;\ C^W_j = C''_{j,1};\ C^P_j = C'_{j,n};$$

$$C^E_{j,i} = C'_{j,i};\ C^G_{j,k} = C''_{j,k},\ j = \overline{1, m},\ i = \overline{1, n-1},\ k = \overline{2, n},$$

the number of independent parameters of multi-flow cascade with the known number of stages and splitting ratios is $n(m+3)-2$.

3. Calculation and cascade optimization

Given cascade feed flows $F_1, F_2, \ldots, F_n$, flows of additional products $E_1, E_2, \ldots, E_{n-1}$ and $G_2, G_3, \ldots, G_n$, concentrations of components in the feed flows $C^F_{j,1}, C^F_{j,2}, \ldots, C^F_{j,n}$, $j = \overline{1, m}$. Then there are left n independent parameters, which can be selected by proceeding to partial flows of components mixture. For each j-component of i-stage the partial flows of feed, product and waste equal to

$$L_{j,i} = L_i C_{j,i};\ L'_{j,i} = L'_i C'_{j,i};\ L''_{j,i} = L''_i C''_{j,i}.$$

It can be shown that the cuts of the partial flows satisfy the equations [12]

$$\varphi_{j,i} = L'_{j,i} / L_{j,i} = \sigma_i q_{j,i} / (1 + \sigma_i q_{j,i}),$$

Where σ_i – the proportion of enrichment ratio in the waste of i-stage to the corresponding value in the product. Parameters σ_i, $i = \overline{1, n}$ do not depend on the component under investigation and can be used when calculating cascade as n independent variables.

On account of the balance of the partial flows of components at cascade sections

$$L''_{j,i} - L'_{j,i-1} = W_{j,i},\ j = \overline{1, m},\quad i = \overline{1, n},$$

Where $W_{i,j}$ – transfer flow of j-component at section before the i-stage, equals to

$$W_{j,i} = WC^W_j - \sum_{l=1}^{i-1} F_l C^F_{j,l} + \sum_{l=1}^{i-2} E_l C^E_{j,l} + \sum_{l=2}^{i} G_l C^G_{j,l}.$$

In these equations $L_{j,0} = 0$ and the sums equal to zero, if the upper summation limits are smaller than the lower ones. Evaluating the partial flows through the sections, that is

$$L_{j,i} = \frac{1}{1 - \varphi_{j,i}} \left(W_{j,i} + \varphi_{j,i-1} L_{j,i-1}\right),$$

can be determined

$$L_{j,i} = \frac{1}{\varphi_{j,i}} \sum_{l=1}^{i} W_{j,l} \prod_{s=l}^{i} \frac{\varphi_{j,s}}{1-\varphi_{j,s}}, \quad j = \overline{1,m}, \quad i = \overline{1,n}.$$

At the product of cascade for each *j*-component the condition should be true

$$L'_{j,n} = L_{j,n}\varphi_{j,n} = \sum_{i=1}^{n} F_i C_{j,i}^{F} - \sum_{i=1}^{n-1} E_i C_{j,i}^{E} - \sum_{i=2}^{n} G_i C_{j,i}^{G} - W_j ,$$

where $W_j = WC_j^W$ – partial flow of the *j*-component in the cascade waste and the equation on the right part shows the partial flow $P_j = PC_j^P$ in product. On rearrangements it follows

$$W_j = \sum_{i=1}^{n} (F_i C_{j,i}^{F} - E_{i-1} C_{j,i-1}^{E} - G_{i+1} C_{j,i+1}^{G}) A_{j,i} / B_j ,$$

where $E_0 = G_{n+1} = 0$, and coefficients $A_{j,i}$ and B_j equal to

$$A_{j,i} = 1 + \sum_{l=i+1}^{n} \prod_{s=l}^{n} \frac{\varphi_{j,s}}{1-\varphi_{j,s}}; \; B_j = 1 + \sum_{l=1}^{n} \prod_{s=l}^{n} \frac{\varphi_{j,s}}{1-\varphi_{j,s}}.$$

This formula helps to calculate the partial flows W_j, $j = \overline{1,m}$ in accordance with the selected stage parameters σ_i, $i = \overline{1,n}$. Its distinction is that the components concentrations in the additional product flows of cascade are not independent and are found during the process of calculation. In this context it is suitable to use the iterative procedure of product $C_{j,1}^{E}, C_{j,2}^{E}, ..., C_{j,n-1}^{E}$, $C_{j,2}^{G}, C_{j,3}^{G}, ..., C_{j,n}^{G}$ and $j = \overline{1,m}$. In such a case the first calculation can be made without additional products and the received values of components' concentrations in the waste and product flows of stages can be accepted as the initial approximations.

When the partial flows W_j are determined, the flows $W_{j,i}$, $L_{j,i}$, $j = \overline{1,m}$, $i = \overline{1,n}$ and other parameters can be calculated:

$P_j = \sum_{i=1}^{n} F_i C_{j,i}^{F} - \sum_{i=1}^{n-1} E_i C_{j,i}^{E} - \sum_{i=2}^{n} G_i C_{j,i}^{G} - W_j$; $W = \sum_{j=1}^{m} W_j$; $P = \sum_{j=1}^{m} P_j$; $C_j^W = W_j / W$; $C_j^P = P_j / P$; $L_i = \sum_{j=1}^{m} L_{j,i}$; $C_{j,i} = L_{j,i} / L_i$ and etc. It should be noted that at the equal parameters σ_i of the stages, the multi-flow cascade is a quasiideal cascade with the random number of feed flows and additional products. In particular case of one feed flow it is *R*-cascade with additional products. But if vary parameters of the stages σ_i and $i = \overline{1,n}$, an optimization of the cascade can be made.

4. Calculation results and arguments

In the modeling experiment on separation of seven-components mixture of molybdenum hexafluoride were considered cascades with one feed flow, one main product, one waste flow and two additional product flows. Component concentrations in the feed flow were considered as the natural [1]: ^{92}Mo – 14.84, ^{94}Mo – 9.25, ^{95}Mo – 15.92, ^{96}Mo – 16.68, ^{97}Mo – 9.55, ^{98}Mo – 24.13, ^{100}Mo – 9.63 %. Quantity of stages in cascades is n = 90, number of feed flow stage is f = 45. Splitting ratio of stages were determined in reliance on the accepted value per unit of difference of mass numbers $q_0 = 1{,}107$. Calculations were made for different cases of ^{95}Mo and ^{96}Mo isotopes concentrations.

Cascades' optimization was carried out by the criterion of minimum of the total flow in providing of $\underline{C_3}$ ^{95}Mo and $\underline{C_4}$ ^{96}Mo preset concentrations:

$$\sum_{i=1}^{n} L_i \to \min, \quad C_3^G \geq \underline{C_3}, \; C_4^E \geq \underline{C_4},$$

where C_3^G, C_4^E – defined at every step of optimization concentrations of components in stages with the additional products. Additional products were calculated respectively from the waste and product stages of cascade with the highest concentrations of ^{95}Mo and ^{96}Mo. Stage numbers were selected in each calculation through consideration of isotope concentration change in triple-flow cascade without additional products.

The optimization problem in worded form is characterized by a wide range of optimized variables and restrictions to concentrations of target components. Direct methods of optimization, conditioned by decision determination on the base of numerical calculations of target functions and developed for unconstrained optimization tasks, are get trapped at the boundaries area of parameters determination. As a consequence, the optimization was carried out in two phases.

At the first stage the cascade was divided into three sections of stages with the identical σ_i, which provided compliance with the limits with some reserve of target components' concentrations. The division into sections was made in accordance with the results of the total flow for several areas with suggested additional products. Selection of σ_i was carried out taking into account their connection with isotope mass numbers by the splitting ratios [12]. At first section was provided the current of enrichment of ^{96}Mo and light isotopes toward the cascade waste flow. At third section ^{95}Mo and heavier isotopes were concentrated toward the cascade waste flow. At second section the currents of enrichment of ^{95}Mo and ^{96}Mo were opposite: the first was concentrated toward the cascade product flow and the second - to the cascade waste flow.

At the second stage was carried out a correction of σ_i on selected steps by the Hooke — Jeeves numerical method [13]. The present method is the simplest one as compared with heuristic algorithms, simulated annealing method and the other well-known methods. It does not provide a search of the absolute minimum of the total flow at limits but gives one of the guaranteed solutions. At the heart of this method is the coordinate-wise exploring search and optimization according to sample. To choose the best solutions the Hooke — Jeeves numerical method was used under a variety of changes paths of stages parameters σ_i, $i = \overline{1,n}$.

The calculations made it clear that in case of concentration of the intermediary components ^{95}Mo and ^{96}Mo, it is typical the distribution of the feed flow along the length of the cascade with two outstanding peaks. They correspond to division of the cascade into segments with the common parameters σ_i, developing around the areas of additional product flows. These peaks are similar to a flow expansion [6–8] but are expressed more tangibly. An additional point is that there is none or little peak in the point of cascade feed flow.

In figure 2 is shown the feed flow distribution and in figure 3 is shown a graph of ^{95}Mo and ^{96}Mo isotope concentration changes through the stages of optimal cascade. In 30-th stage of additional product flow of such cascade is produced isotope ^{96}Mo and in 72-nd - isotope ^{95}Mo with the preset concentration of 67 %. Calculations were carried out at the identical flows $E = G$ and proportions $E / P = 0.595$ and $E / W = 0.335$. Initial values of parameter σ_i were equal to 0.76 in the section from the 1-st up to 30-stages, 0.64 from 31-st up to 75-th stages and 0.42 from the 76-th up to 90-th stages. They showed the initial concentrations for both isotopes as 67.4 % and they determined, to a great extent, the feed flows through the stages.

Molybdenum components mass content presented in the outflows of the cascade is shown in table 1. Concentration of the lightest isotope ^{92}Mo in the main product is 57 %, concentration of isotope ^{98}Mo in waste flow is – 50.8 %. It shows that it is possible to receive four highly concentrated components in the cascade (table 1).

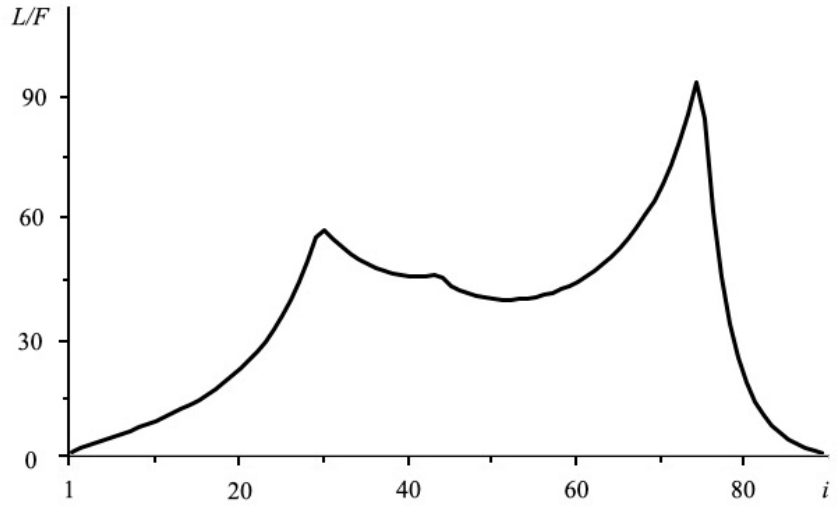

Figure 2. Distribution of feed flows through the cascade stages, optimized for receiving the ^{95}Mo and ^{96}Mo concentrated isotopes.

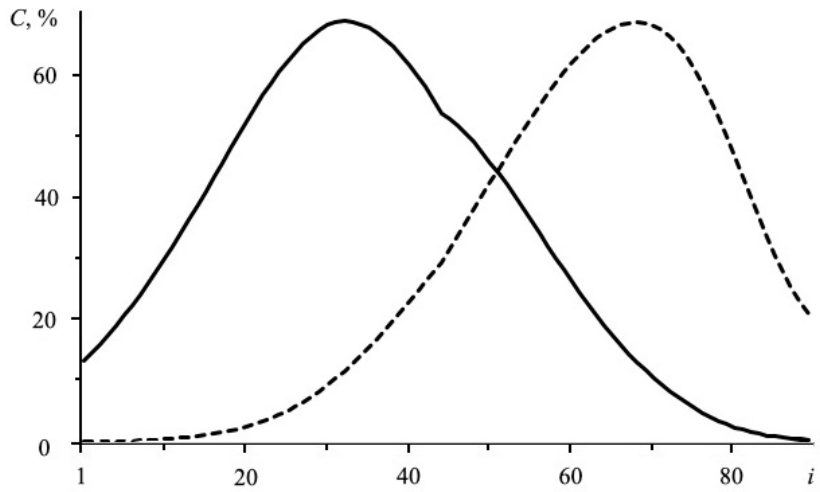

Figure 3. ^{96}Mo (—) isotope concentration change in product flow and ^{95}Mo (---) isotope concentration change in waste flow of stages for optimal cascade.

Table 1. Concentrations of molybdenum components in the flows of optimal cascade for concentration in the additional product flows ^{95}Mo and ^{96}Mo ($E = G$, $E / P = 0.595$ and $E / W = 0.335$), %.

Parameter	^{92}Mo	^{94}Mo	^{95}Mo	^{96}Mo	^{97}Mo	^{98}Mo	^{100}Mo
Feed flow *F*	14.84	9.25	15.92	16.68	9.55	24.13	9.63
Waste flow *W*	$2.2\cdot10^{-8}$	$1.9\cdot10^{-4}$	0.12	10.95	17.03	50.75	21.15
Product *E*	$7.5\cdot10^{-3}$	0.22	9.53	67.01	12.78	9.39	1.06
Product *G*	3.03	19.23	67.00	10.66	0.07	$4.5\cdot10^{-3}$	$4.4\cdot10^{-6}$
Product *P*	57.02	25.10	17.38	0.51	$5.4\cdot10^{-4}$	$6.2\cdot10^{-6}$	$1.6\cdot10^{-10}$

Flow augmentation of additional products leads at the same initial values of σ_i and sections of their determination to a dilution which can be provided at optimization. In Table 2 as an example are shown the calculation results of the optimal cascade with flows ratio of $E / P = 0.812$ and $E / W = 0.430$. The obtained ^{96}Mo isotope concentration in the additional product in 30-th stage is 62.7 % and ^{95}Mo isotope concentration in the additional product in 72-nd stage is 62.5 %. In addition to that the total flow had decreased by 24 %. Concentrations of ^{92}Mo in the main product increased by 63.1 % and ^{98}Mo in the waste flow of cascade – 52.7 %.

Table 2. Concentrations of molybdenum components in the flows of optimal cascade for concentration in the additional product flows ^{95}Mo and ^{96}Mo ($E = G$, $E / P = 0.812$ and $E / W = 0.430$), %.

Parameter	^{92}Mo	^{94}Mo	^{95}Mo	^{96}Mo	^{97}Mo	^{98}Mo	^{100}Mo
Feed flow *F*	14.84	9.25	15.92	16.68	9.55	24.13	9.63
Waste flow *W*	$2.3\cdot10^{-8}$	$1.9\cdot10^{-4}$	0.10	8.38	16.40	52.67	22.45
Product *E*	0.01	0.28	9.12	62.68	14.89	11.67	1.34
Product *G*	4.67	22.77	62.50	9.98	0.07	$4.7\cdot10^{-3}$	$3.7\cdot10^{-6}$
Product *P*	63.11	23.00	13.47	0.42	$5.7\cdot10^{-4}$	$6.8\cdot10^{-6}$	$1.7\cdot10^{-10}$

Table 3. Concentrations of molybdenum components in the flows of optimal cascade for concentration in the additional product flows ^{95}Mo and ^{96}Mo ($E = G$, $E / P = 0.427$ and $E / W = 0.251$), %.

Parameter	^{92}Mo	^{94}Mo	^{95}Mo	^{96}Mo	^{97}Mo	^{98}Mo	^{100}Mo
Feed flow *F*	14.84	9.25	15.92	16.68	9.55	24.13	9.63
Waste flow *W*	$2.7\cdot10^{-8}$	$2.5\cdot10^{-4}$	0.18	14.40	17.15	48.38	19.89
Product *E*	$4.9\cdot10^{-3}$	0.19	9.83	70.01	11.10	7.98	0.88
Product *G*	2.75	17.13	70.01	10.05	0.05	$3.6\cdot10^{-3}$	$3.5\cdot10^{-6}$
Product *P*	51.62	25.52	22.24	0.62	$5.8\cdot10^{-4}$	$7.3\cdot10^{-6}$	$2.1\cdot10^{-10}$

Flow reduction of additional products has the opposite results (table 3). Correlating $E / P = 0.427$ and $E / W = 0.251$ the received concentrations of ^{96}Mo isotope in the additional product of 29-th stage and ^{95}Mo in 72-nd stage are 70 %. The total flow had increased as opposed to the example, taken from Table 1, by 11 %. The concentrations of ^{92}Mo in the main product have decreased by 51.6 % and the concentrations of ^{98}Mo in the waste flow of cascade – by 48.4 %.

5. Conclusion

The worked out method makes possible an optimization of cascades with additional product flows, wherein are collected components, which are intermediary by mass number. Calculated cascades have the smallest total flow at preset concentrations of target isotopes.

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